

# FUGITIVE METHANE EMISSIONS FROM TWO EXPERIMENTAL BIOCOVERS CONSTRUCTED WITH TROPICAL RESIDUAL SOILS: FIELD STUDY USING A LARGE FLUX CHAMBER

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## ABSTRACT

This study aimed at assessing the response of two experimental passive methane oxidation biocovers (PMOB) installed in a Brazilian landfill located in Guarapuava, State of Paraná. The PMOBs covered an area of 18 m<sup>2</sup> each, and were 0.70-m-thick. The first PMOB (control subarea) was constructed using the same soil used to cover closed landfill cells, i.e. a typical residual soil. The second PMOB (enriched subarea) was constructed with a mixture of the residual soil and mature compost, with a resulting organic matter content equal to 4.5%. CH<sub>4</sub> and CO<sub>2</sub> surface fluxes were measured in a relatively large (4.5 m<sup>2</sup>) static chamber. CH<sub>4</sub>, CO<sub>2</sub> and O<sub>2</sub> concentrations were also measured at different depths (0.10, 0.20, 0.25 and 0.30 m) within PMOBs. The concentrations from the raw biogas were also measured. Methane oxidation efficiencies (Eff<sub>ox</sub>) were estimated based on the CO<sub>2</sub>/CH<sub>4</sub> ratio. The average CH<sub>4</sub> and CO<sub>2</sub> concentrations in the raw biogas (42% and 32%, respectively) for the 16 campaigns corroborated those typically found in Brazilian landfills. Lower CH<sub>4</sub> fluxes were obtained within the enriched subarea (average of 20 g.m<sup>-2</sup>.d<sup>-1</sup>), while the fluxes in the control subarea averaged 34 g.m<sup>-2</sup>.d<sup>-1</sup>. Eff<sub>ox</sub> values averaged 42% for the control subarea and 80% for the enriched one. The results indicate that there is a great potential to reduce landfill gas (LFG) emissions by using passive methane oxidation biosystems composed of enriched substrates (with a higher content of organic matter).

## 1. INTRODUCTION

In Brazilian municipal solid waste (MSW) landfills, where there is no any biogas active recovery system in most of them, a common practice to reduce biogas emissions is to install a vertical drain – usually constructed with very coarse gravel – and flare it. These drains, which are often operated manually, are submitted to the large settlements that usually take place in landfills. Consequently, the integrity of the usually poorly maintained drains is negatively affected, reducing their capacity to drain landfill gas (LFG). In addition, Brazilian landfill covers are often constructed with readily available materials, which are placed in one single layer. Considering these premises, it can be expected that a significant proportion of the generated LFG migrates through the cover system in the form of fugitive emissions (Maciel and Jucá, 2011).

Several studies have shown that a passive biosystem (where the biogas passes through the cover naturally, without pumping it) installed in the cover system (interim or fi-

nal) can be a very effective complement to active systems in reducing fugitive emissions of methane and odorous substances (Abichou et al., 2006a; Cabral et al., 2010a; Capanema et al., 2013; Capanema et al., 2014; Geck et al., 2016; Lucernoni et al., 2016; Roncato and Cabral, 2012; Sadasivam and Reddy, 2014; Scheutz et al., 2009). Most of these studies documented the performance of passive biosystems in temperate climates, while (to the authors' knowledge) no field-scale studies have been performed to document the performance of passive biosystems in Brazil (a tropical country) and employing residual soils. Such passive systems are in great need in developing countries due to the almost complete absence of active systems.

Documentation of methane, odorous substances and organic compound emissions has often relied on the use of flux chambers that cover surfaces lower than 1.0 m<sup>2</sup> (Abichou et al., 2006a,b; Gallego et al., 2014; Hudson and Ayoko, 2008; Scheutz et al., 2009; Trégourès et al., 1999). The low cost and simplicity of the static chamber



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method resulted in its widespread use. However, given the fact that fugitive emissions are often concentrated in cracks that cover very small subareas (e.g. Geck et al., 2016; Rachor et al., 2011; Rachor et al., 2013), in addition to varying strongly in time, chances are that concentrated fluxes in these cracks will be missed during emissions assessments using small flux chambers. According to Geck et al. (2016), extrapolation of total emissions from measurements performed with small flux chambers can be questionable.

The present study documented the response of two experimental passive methane oxidation biosystems (PMOBs) installed in a Brazilian landfill where biogas is vented out passively. The landfill is located in the sub-tropical subarea of central Paraná, a southern state. The first PMOB (control) was constructed with the same residual soil that has been employed as both interim and final cover by the landfill operator. The other PMOB was constructed with this same soil after being enriched with organic-matter-rich compost. This study focused on the capacity of the two PMOBs to oxidize methane and on the magnitude of surface emissions. Oxidation efficiency was assessed by means of the  $\text{CO}_2$  to  $\text{CH}_4$  ratio along several gas profiles (Gebert et al., 2011a).

For this study, we designed and constructed a low-cost and easy-to-build large-scale chamber. Table 1 presents the characteristics of several flux chambers used in recently reported studies about landfill biogas emissions. Flux chambers with surfaces up to 17.7 m<sup>2</sup> and 880-L volume have been reported. Ideally, according to Rochette and Eriksen-Hamel (2008), the surface covered by the chamber and its volume should be proposed as a function of flux intensity (or biogas loading). Given the fact that the PMOBs were installed directly over the waste mass, and we did not have access to flux intensity data, our design was based on the largest possible chamber we could build and carry.

## 1.1 Abbreviations

AVG: Average  
 Eff<sub>ox</sub>: Methane oxidation efficiencies  
 f: Biogas mass flux  
 FID: Flame ionization detector  
 GC: Gas chromatography  
 LFG: Landfill gas  
 MSW: Municipal solid waste  
 OM: Organic matter  
 PMOB: Passive methane oxidation biocover  
 STP: Standard temperature and pressure  
 TCD: Thermal conductivity detector  
 TOC: Total organic carbon

## 2. MATERIAL AND METHODS

### 2.1 Characterization of the study area and the cover soil

This study was carried out in a landfill located in Guarapuava, a city located in the south-central region of the State of Paraná (Brazil). This landfill has been used to dispose of non-hazardous municipal solid wastes (MSW). The authors selected a region within the landfill as flat as possible and as far away as possible from vertical drains, in order to avoid preferential pathways of the biogas to these drains, cracks on the surfaces and ensure that  $\text{CH}_4$  was present in measurable concentrations at the surface. The cover in the selected area had been placed 4.5 years before beginning of our experiments. Chamber measurements were then performed to assess  $\text{CH}_4$  emissions.

Within this area, two 18 m<sup>2</sup> subareas were delimited: one called the “control subarea” and the other “enriched subarea” (with compost), covered by a 0.70-m-thick residual soil layer. The spacing between each subarea was 1.0 m. The cover soil in the control subarea was the same as else-

**TABLE 1:** Shapes and sizes of flux chambers reported in the literature.

Shape of the chamber	Dimensions (m)		Surface (m <sup>2</sup> )	Volume (L)	Flux g.m <sup>-2</sup> .d <sup>-1</sup>	References
	Base	Height				
Rectangular	0.65	0.28	0.42	120	N.R. <sup>1</sup>	Chanton and Liptay (2000)
Rectangular	0.63	0.20	0.40	80	330 – 596 18.1 – 117.5 Chanton et al. (2011) N.R. <sup>1</sup>	Abichou et al. (2006b); Stern et al. (2007); Chanton et al. (2011)
Rectangular	1.80 (L) x 1.20 (W)	0.25	2.10	504	330-596 (maximum values) 18.1-117.5 (mean values) Chanton et al. (2011) N.R. <sup>1</sup>	Capanema et al. (2013), Lakhout et al. (2014)
Rectangular	1.22 (L) x 0.76 (W)	0.25	0.93	241	N.R.*	Capanema et al. (2014)
Quadratic	N.R. <sup>1</sup>	N.R. <sup>1</sup>	1.00	50	5.0 – 389.2	Araujo and Ritter (2016)
Quadratic	0.50 (L and W)	0.10	0.25	25	N.R. <sup>1</sup>	Lucernoni et al. (2016)
Quadratic	0.40 (L and W)	0.05	0.16	0.008	N.R. <sup>1</sup>	Monteiro et al. (2016)
Quadratic	4.2 (L and W)	0.50	17.7	880	0.98-6.69	Geck et al. (2016)
Rectangular	3.00 (L) x 1.5 (W) <sup>2</sup>	0.2	4.50	900		This study

<sup>1</sup> N.R.: not reported; <sup>2</sup> divided into 2 sections for ease of transportation. The sections are assembled on site.

where in the site (sandy clayey silt, 86.3% sieved through a 200-Mesh sieve); its average organic matter content is equal to 0.4%. In the enriched subarea, the uppermost 0.15 m of soil was substituted by a mixture of the mature compost (originally with 32% organic matter content) and the same natural soil, resulting in a substrate with 81.4% sieved through a 200-Mesh sieve and an organic matter content equal to 4.5%. The intention was to foster bacterial growth and assess addition of nutrients in order to improve passive methane oxidation. The pH of the soil (~6.6 for the control subarea; and 7.1 for the enriched subarea) varied very little throughout the study period. It can therefore be assumed that pH would not constitute a limiting parameter in this study. Moreover, it is known that methane-oxidizing bacteria perform better in pH near to neutrality (Delhomnie and Heitz, 2005).

The Atterberg limits determined for the control subarea were: Liquidity limit = 51%, Plasticity limit = 36%; and the Plasticity Index was 15%. The values found for the enriched subarea were: Liquidity limit = 51%, Plasticity limit = 36%; and the Plasticity Index was 16%. For the control subarea, the Standard Proctor's optimum moisture was 29.6% for a maximum dry density equal to 1.47 g/cm<sup>3</sup>, whereas the respective values for the enriched subarea were 35.6% and 1.39 g/cm<sup>3</sup>.

## 2.2 Characterization of raw biogas

Assessment of the characteristics of the raw biogas was performed by installing raw biogas pipes in each of the two subareas. Stainless steel pipes with a diameter of 10 mm were buried at a depth of 1.0 m in order to reach the interior of the waste mass. Once it was not possible to measure the loading rate or even the flow rate of biogas through the biocovers, the raw biogas concentration was the only parameter monitored, in order to determine the ox-

idation efficiency of the cover layer in both of the evaluated subareas.

The concentration of the main gases that compose the raw biogas was determined with the aid of a Columbus portable gas analyzer (Columbus Instruments Inc.) equipped with infrared sensors for detecting CO<sub>2</sub> and CH<sub>4</sub> in a range of 0-100 vol.%, and coupled to an electrochemical sensor for detecting O<sub>2</sub> between 0-21 vol.%. The measurement accuracy for methane and carbon dioxide is about 2% and 1% of the value read for oxygen. The biogas was sampled from the pipes using a 60-mL syringe and injected into the gas analyzer. N<sub>2</sub> concentrations were calculated by subtracting the sum of CO<sub>2</sub>, CH<sub>4</sub> and O<sub>2</sub> concentrations from 100% (simplifying assumption).

## 2.3 Assessment of surface emissions using the large flux chamber

### 2.3.1 Flux chamber specifications

A flux chamber was built in a similar way to those used by Capanema et al. (2013) and Lakhout et al. (2014); as shown in Figure 1a. It covers an area equal to 4.5 m<sup>2</sup> with a total internal volume equal to 0.9 m<sup>3</sup>.

A fishbone-shaped system with perforated copper piping was installed in the interior part of the flux chamber, in order to capture the gas from all points inside the chamber, directing them to the exit point (sampling point - Figure 1b). The frame (onto which the cover of the flux chamber rested) was inserted at 0.15 m depth and sealed with bentonite, as recommended by Capanema et al. (2014). The top of the frame had a groove where the water was poured to prevent the entry of atmospheric air during flux measurements. Each subarea had its own fixed frame.

Inside the chamber, two small battery-operated fans were installed to ensure homogenization of the emitted biogas, according to the field recommendations applied



**FIGURE 1:** Biogas samplings: (a) flux chamber; (b) biogas sampling point.



by Abichou et al. (2006b), Scheutz et al. (2014) and Stern et al. (2007). A mercury thermometer was also installed to measure the temperature variation inside the chamber, and thus determine the biogas concentrations according to the standard temperature and pressure (STP).

Water content probes (ECH<sub>2</sub>O CE-5 sensor, Decagon, Pullman, USA) were installed at 0.15 m below surface in both subareas to determine the soil water content. All samples were collected respecting at least two days between the last precipitation and the sampling campaign. However, there were intense rains in the days leading up to some campaigns, considerably increasing the soil moisture content. Precipitation and atmospheric pressure data were obtained from the Weather System of Parana State database, because site-specific data was not available.

### 2.3.2 Sampling and analysis of CH<sub>4</sub> and CO<sub>2</sub> emissions

Sixteen campaigns were carried out to determine CH<sub>4</sub> and CO<sub>2</sub> fluxes using the chamber. Emitted biogas samples were collected at 2-minute intervals using a 60 mL syringe (Figure 1b). All samples were collected within one hour, as suggested by the UK Environment Agency (2010).

The samples were inserted into 30 mL glass vials that had been previously placed under vacuum, and then sealed with a septum "crimp" (Du et al., 2006; Jantalia et al., 2008). They were subsequently analyzed by GC-flame ionization-thermal conductivity using a Shimadzu FID-TCD, equipped with a 5-m long Carboxen 1000 packed column (60/80 mesh). The main testing parameters are as follows: oven heating ramp: 40°C (6 min); heating rate: 20°C/min up to 220°C; carrier gas: argon. Gas chromatography analysis were only required for methane concentrations below 1% (below the Columbus detection limit).

### 2.3.3 Surface methane mass flux calculations

Biogas mass fluxes at the surface were calculated based on the results obtained using Equation 1 (according to Cabral et al., 2010a and Perera et al., 2002), corrected for the standard temperature and pressure (STP).

$$f = (V/A) \times (\Delta C / \Delta t) \times (273,15 / (273,15 + T_{int})) \times (P_{atm} / 1013) \quad (1)$$

where: V = internal volume of the flux chamber (m<sup>3</sup>); A = cover layer area of the chamber (m<sup>2</sup>);  $\Delta C / \Delta t$  = represents the slope of the plot relating the change in gas concentration to time (mg.m<sup>-3</sup>.s<sup>-1</sup>); T<sub>int</sub> = internal temperature of the gas in the chamber (°C); P<sub>atm</sub> = atmospheric pressure (mbar).

## 2.4 Vertical concentration profiles of biogas

### 2.4.1 Installation of the gas probes

Five nests of stainless steel gas probes were placed in each subarea, as shown in Figure 2. In each nest, the 10-mm (internal diameter) gas probes were inserted using a metal auger to desired position (0.10, 0.20, 0.25 and 0.30 m) below the surface, following the methodology described by Cabral et al. (2010a). After insertion, the upper ends were capped with a rubber septum.

### 2.4.2 Determining gas concentrations

Concomitantly with the 16 flux measurements, campaigns were also performed for determining the gas concentrations along the cover layer of the two evaluated subareas. First, each gas probe was purged of the volume of air initially contained therein using a 60 mL syringe. After one hour, a biogas sample was collected and analyzed in the portable gas analyzer "Columbus". Again, gas chromatography analysis were only required for methane concentrations below the Columbus detection limit ( $\approx 1\%$ ).

## 2.5 Estimation of methane oxidation efficiency (Eff<sub>ox</sub>) based on gas profile data

The share of oxidized methane (x) at a certain depth was determined in both subareas, according to Equation 2 (Gebert et al., 2011b).

$$(CO_{2LFG} + x) / (CH_{4LFG} - x) = CO_{2i} / CH_{4i} \quad (2)$$

where x = share of oxidized CH<sub>4</sub> (vol.%), CH<sub>4LFG</sub> = CH<sub>4</sub> concentration of the landfill gas (vol.%), CO<sub>2LFG</sub> = CO<sub>2</sub> concentration of the landfill gas (vol.%), CH<sub>4i</sub> = CH<sub>4</sub> concentration at depth i (vol.%), CO<sub>2i</sub> = CO<sub>2</sub> concentration at depth "i" (vol.%).

The ratio between the percentage of oxidized methane at the depth of 0.10 m and the methane concentration in the raw biogas (CH<sub>4LFG</sub>) determines the methane oxidation efficiency (Eff<sub>ox</sub>) in % according to Equation (3) (Gebert et al., 2011b). One important hypothesis associated with this method relates to soil respiration. The amount of CO<sub>2</sub> produced by soil respiration needs to be negligible compared to the CO<sub>2</sub> produced due to methane oxidation (Geck et al., 2016). It is assumed herein that respiration is negligible. We base this assumption on results presented by Gebert et al. (2011b), who found, in a batch experiment using soil with total organic carbon (TOC) 4.9%-7.5%, that

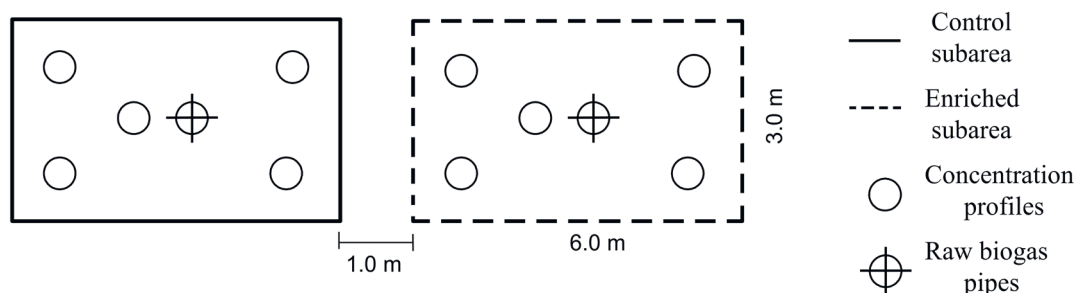


FIGURE 2: Concentration profiles of the subareas (enriched and control).

CO<sub>2</sub> respiration accounted for less than 2% of the observed CO<sub>2</sub> production. In the same study, the oxidation efficiency was only slightly overestimated when a soil with 6% organic matter was tested.

$$Eff_{ox} = (x/CH_{4-LFG}) \times 100 \quad (3)$$

where x = share of oxidized CH<sub>4</sub> (vol.%), CH<sub>4-LFG</sub> = methane concentration in the landfill gas (vol.%).

### 3. RESULTS AND DISCUSSION

#### 3.1 Raw biogas characterization

The concentration values of CH<sub>4</sub>, CO<sub>2</sub> and O<sub>2</sub> in the raw biogas are shown in Figure 3. Raw biogas samples were collected at a depth of 1.0 m below the surface. The average (AVG) CH<sub>4</sub> concentration in the raw biogas for the 16 campaigns was 42% (with standard deviation, σ equal to ±4.49%). For CO<sub>2</sub>, the AVG concentration was 32% (σ=±2.53). The concentration ranges in Figure 3 are typical of Brazilian landfills (Audibert and Fernandes, 2013; Can-

diani et al., 2011). The presence of O<sub>2</sub> in the waste is not a surprise. Despite near optimal conditions for accelerated waste degradation, cover systems in most landfills in Brazil (and in the developing world) are often composed of a single layer of soil; cracks can be formed during dry periods, thereby allowing penetration of atmospheric air. In fact, O<sub>2</sub> penetration has been documented for landfills in several subareas of the globe, including the UK (Barry et al., 2003), Iceland (Kjeld et al., 2014), Australia (Obersky et al., 2018) and Brazil (Audibert and Fernandes, 2013).

#### 3.2 Evaluation of the surface emissions

Surface methane mass flux and soil temperatures (at 0.10 m depth) throughout the 16 campaigns and in each subarea are shown in Figure 4.

Methane fluxes in the control subarea ranged between 0 and 53 g.m<sup>-2</sup>.d<sup>-1</sup> (AVG=34 g.m<sup>-2</sup>.d<sup>-1</sup> and σ=±13.5), whereas methane fluxes in the enriched subarea varied between 0 and 49 g.m<sup>-2</sup>.d<sup>-1</sup> (AVG=20 g.m<sup>-2</sup>.d<sup>-1</sup> and σ=±15.3). On Feb 22<sup>nd</sup>, the soil was too wet and there was no noticeable

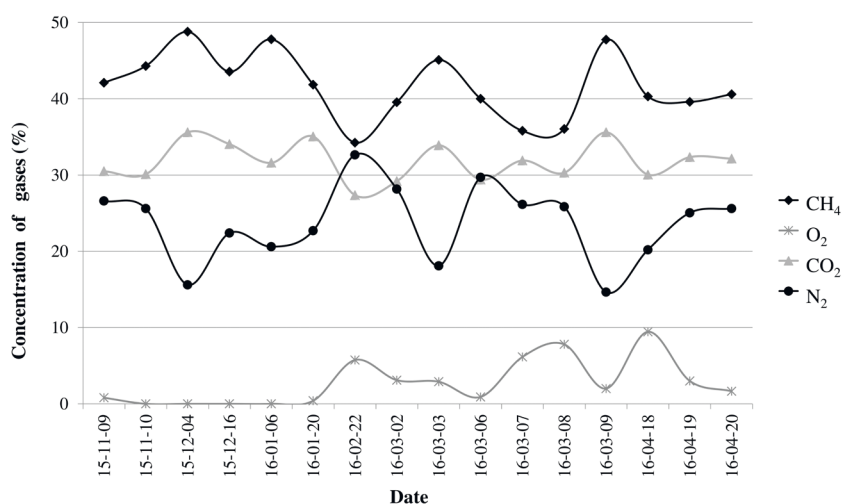


FIGURE 3: Average concentration of raw biogas throughout the 16 campaigns.

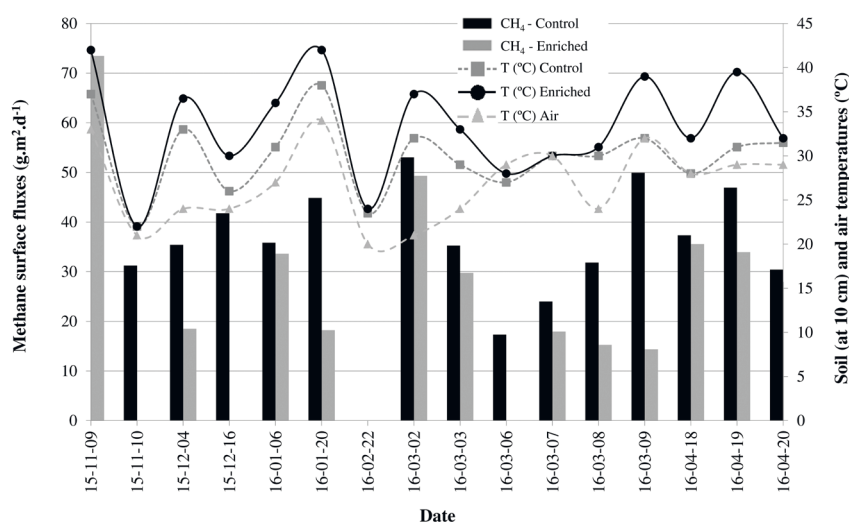


FIGURE 4: Surface methane mass fluxes (g.m<sup>-2</sup>.d<sup>-1</sup>), air and soil temperatures for both subareas.

increase in gas concentrations inside the flux chamber over several hours. Consequently, fluxes were considered equal to zero in both zones for this particular date. It is noteworthy that  $\text{CH}_4$  fluxes in the enriched subarea were always lower or much lower than in the control subarea, with the exception of the first field campaign (on Nov. 9<sup>th</sup>, 2015) in the enriched subarea, when the microorganisms were probably acclimatizing and adapting to the environment.

Abichou et al. (2009) also compared an organic-rich biocover with an organic-poor cover and observed much lower  $\text{CH}_4$  fluxes from the organic-rich biocover. The slightly lower average methane flux on the surface of the compost-enriched subarea may be associated with the greater organic matter content, which created better conditions for the development of ubiquitous methanotrophic bacteria (Humer and Lechner, 2001). An in-depth study using modern microbiology analysis tools could not be performed to confirm the preceding assertion.

The air temperature varied between 20 and 33°C during the 16 campaigns, while inside the interim cover soils temperatures varied between 22 and 38°C (control) and between 22 and 42°C (enriched), respectively. The higher soil temperatures within the enriched subarea (Figure 4) probably resulted from more intense microbial activity. Carbon dioxide mass fluxes are shown in Figure 5.

In the control subarea,  $\text{CO}_2$  fluxes vary between 33 and 721  $\text{g.m}^{-2}.\text{d}^{-1}$  ( $\text{AVG}=251 \text{ g.m}^{-2}.\text{d}^{-1}$ ;  $\sigma=\pm 168$ ), while in the enriched subarea they vary between 0 and 794  $\text{g.m}^{-2}.\text{d}^{-1}$  ( $\text{AVG}=316 \text{ g.m}^{-2}.\text{d}^{-1}$ ;  $\sigma=\pm 245$ ). Fernandes (2009) reported  $\text{CO}_2$  fluxes in Brazilian landfills between 0 and 388  $\text{g.m}^{-2}.\text{d}^{-1}$ , and between 29 and 233  $\text{g.m}^{-2}.\text{d}^{-1}$  for conventional and enriched (soils), respectively.

The carbon dioxide surface flux is higher than the methane flux in the enriched area for most of the assays (e.g. data from January 20<sup>th</sup> and March 9<sup>th</sup>, 2016 in Figure 4 and Figure 5), highlighting the higher conversion of methane to carbon dioxide and water. A similar trend was reported

by Christophersen et al. (2001), who found carbon dioxide emissions of 90  $\text{g.m}^{-2}.\text{d}^{-1}$ , slightly higher than those found for methane (75  $\text{g.m}^{-2}.\text{d}^{-1}$ ). Scheutz et al. (2011) verified this increase in carbon dioxide emission in lysimeters with mature and stable compost.

Despite  $\text{CH}_4$  emissions being considered equal to zero on Feb. 22<sup>nd</sup>, 2016, due to wet conditions,  $\text{CO}_2$  concentration increases were measurable in the two zones for this same date. It can only be speculated that minimal soil respiration near the surface led to these non-zero values.

### 3.3 Vertical profiles of biogas

An average of the biogas composition for all set of gas probes along vertical profiles in the first 30 cm of the cover layer for both control (Figure 6a) and enriched (Figure 6b) subareas are presented.

The fact that the  $\text{CH}_4$  and  $\text{CO}_2$  curves cross at different depths (0.13 m for control subarea – Figure 6a - and 0.23 m for enriched subarea – Figure 6b) indicates a previous methane oxidation in the enriched subarea and, therefore, a highest capacity of oxidation of this substrate.

Figure 6 also shows that oxygen was present throughout the profile. The average percentage of oxygen remained between 12.4% (control subarea) and 13.9% (enriched subarea). According to Czepiel et al. (1996); Gebert et al. (2003); Jugnia et al. (2008), an oxygen concentration >3% is sufficient for the oxidation reaction. Therefore, oxygen was never a limiting parameter for methanotrophic activity. In addition, it can be observed that  $\text{O}_2$  concentrations decreased in the profile due to soil retention and microbial methane oxidation.

The degrees of saturation were calculated using volumetric data of water content. The degree of saturation is between 71% (control area) and 80% (enriched area). According to Huber-Humer and Lechner (2003), the ideal degree of saturation for methanotrophic activity is between 40 and 80%. The degrees of saturation always remained lower than 85%, which approximately corresponds to the

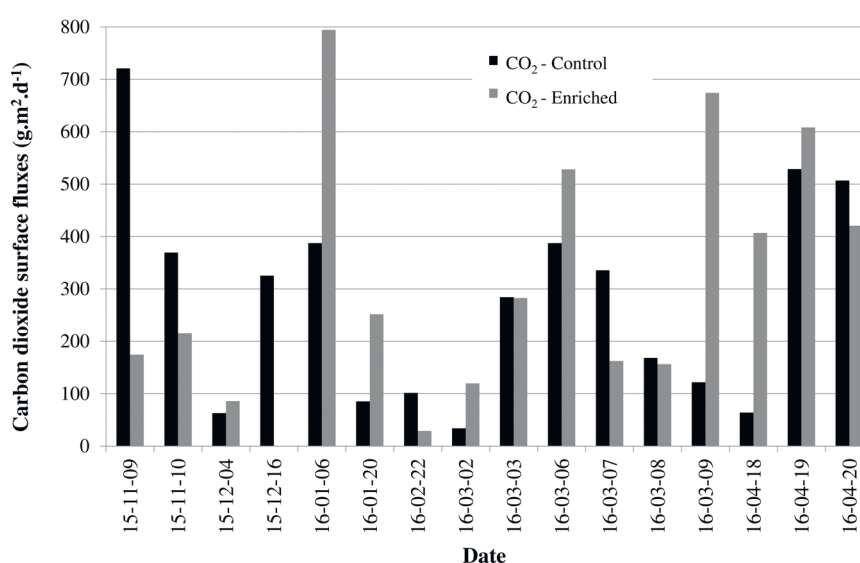


FIGURE 5: Surface carbon dioxide mass fluxes ( $\text{g.m}^{-2}.\text{d}^{-1}$ ) for both subareas.

degree of saturation value beyond which air becomes occluded in the pores of fine grained soils (Nagaraj et al., 2006), such as the one used to construct the experimental covers. Consequently, with the exception of the near-zero  $\text{CH}_4$  flow observed on Feb 22<sup>nd</sup>, no impedance to flow was observed. It is also worth pointing out that when soil moisture content increases (as observed on Feb 22<sup>nd</sup>), the upward flow of  $\text{CH}_4$  and the downward flow of  $\text{O}_2$  in the oxidation layer become limited to diffusion in the liquid phase, delaying the oxidation process (Cabral et al., 2010b).

### 3.4 Methane oxidation efficiencies

Figure 7 presents the average oxidation efficiencies of methane ( $\text{Eff}_{\text{ox}}$ ) for the control and enriched subareas, calculated by Equation (3).

Figure 7 shows that the methane oxidation capacity varied over time and was affected by the type of material constituting the cover soil. Indeed, the methane oxidation efficiencies of the enriched subarea were always high-

er than in the control subarea. The mean efficiencies of methane oxidation at 0.10 m were 42% ( $\pm 1.56\%$ ) and 80% ( $\pm 1.88\%$ ) for the control and enriched subareas, respectively. The average  $\text{CH}_4$  surface emissions were 34 and 20  $\text{g} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$ , respectively.

These results corroborate findings by several other studies. For example, Abichou et al. (2009) reported maximum methane efficiency equal to 63% for the control subarea in a landfill in Florida, while in the biocover it was 100%. Rose et al. (2012) obtained a maximum  $\text{CH}_4$  oxidation efficiency of 67% for a conventional cover layer, while improving it with compost led to a maximum of 97%. Capanema et al. (2013) found a methane oxidation efficiency of 95.8% in a biocover whose O.M. content was similar to the top soil in the enriched subarea reported in the present study. Einola et al. (2008) observed that 96% of the methane was oxidized near the surface, where the topsoil was richer in organic matter.

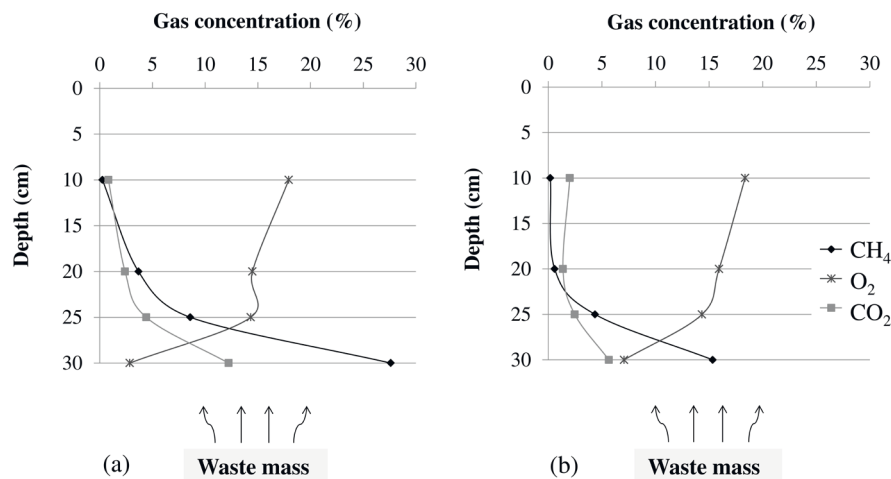


FIGURE 6: Vertical composition profiles of biogas for control (a) and enriched (b) subareas.

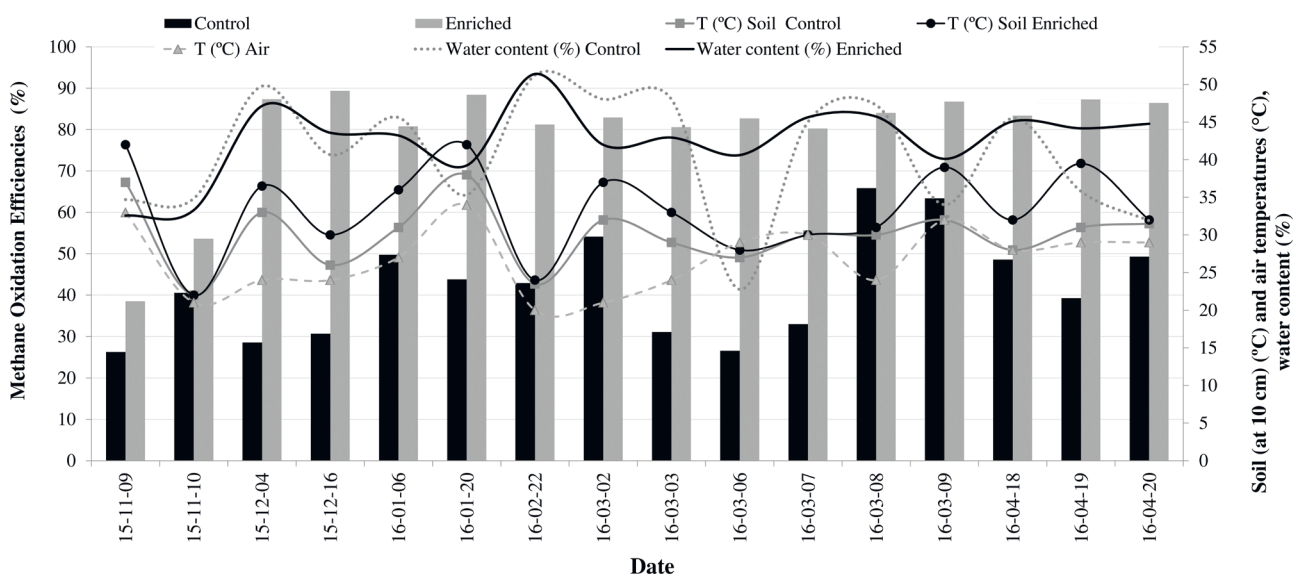


FIGURE 7: Methane oxidation efficiencies ( $\text{Eff}_{\text{ox}}$ ) in the control and enriched subareas.



## 4. CONCLUSIONS

This study reported the capacity of two experimental passive methane oxidation biosystems (PMOBs) consisting of tropical soils typically found in developing nations. One of them was supplemented with organic-rich material (enriched subarea). In general, the average  $\text{CH}_4$  and  $\text{CO}_2$  concentrations in the raw biogas (42% and 32%, respectively) for the 16 campaigns corroborated those typically found in Brazilian landfills. The presence of oxygen in raw biogas resulted from atmospheric air penetration in the thin, single-layer cover, mainly through cracks.

The methane oxidation capacity was quite high for both subareas (control and enriched). Oxidation efficiencies (at a depth of 0.10 m) averaged 42% for the control subarea and 80% for the enriched area.  $\text{CH}_4$  and  $\text{CO}_2$  surface fluxes averaged  $20 \text{ g.m}^{-2}.\text{d}^{-1}$  and  $316 \text{ g.m}^{-2}.\text{d}^{-1}$  in the organic-matter-enriched subarea during the monitoring period, while those measured in the control subarea averaged  $34 \text{ g.m}^{-2}.\text{d}^{-1}$  and  $251 \text{ g.m}^{-2}.\text{d}^{-1}$ , respectively. It is noteworthy that the surface fluxes were obtained using a custom-made  $4.5\text{-m}^2$  flux chamber, which allows for better representativeness of surface fluxes, because it allows inclusion of cracks and other imperfections that may affect measurements.

The lower  $\text{CH}_4$  fluxes and higher oxidation efficiency in the enriched subarea can be associated with the greater organic matter content in the enriched subarea, which created more favourable conditions for the development of ubiquitous methanotrophic colonies (Humer and Lechner, 2001). Temperature conditions, which ranged from 20 to  $42^\circ\text{C}$  at the surface and within the first 10 cm of the cover, favoured methane oxidation.

The results obtained in this study point to the great potential in reducing residual LFG emissions by landfills located in developing nations using low-cost PMOBs constructed with typical tropical soils.

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## REFERENCES

Abichou T, Chanton J, Powelson D, Fleiger J, Escoriza S, Lei Y, Stern J (2006a) Methane flux and oxidation at two types of intermediate landfill covers. *Waste Management*, 26:1305-1312.

Abichou T, Powelson D, Chanton J, Escoriza S, Stern J (2006b) Characterization of methane flux and oxidation at a solid waste landfill. *Journal of Environmental Engineering*, 132:220-228.

Abichou T, Mahieu K, Yuan L, Chanton J, Hater G (2009) Effects of compost biocovers on gas flow and methane oxidation in a landfill cover. *Waste Management*, 29:1595-1601.

Araujo TT, Ritter E (2016) Evaluation of biogas emissions from layers of a landfill cover. *Perspectivas Online*, 16:34-49 (in Portuguese).

Audibert JL, Fernandes F (2013) Preliminary qualitative and quantitative assessment of gases from biodegradation of solid wastes in the landfill of Londrina, Paraná State, Brazil. *Acta Scientiarum Technology*, 35:45-52.

Barry DL, Smith R, Gregory RG, Harries C (2003) Methane Production, Emission and Control during MSW Landfilling. In: *Proceedings of the Ninth International Landfill Symposium*, Cagliari, Sardinia, 6-10 October 2003.

Cabral AR, Moreira JFV, Jugnia L-B (2010a) Biocover Performance of Landfill Methane Oxidation: Experimental Results. *Journal of Environmental Engineering*, 136:785-793.

Cabral AR, Létourneau M, Yanful E, Song Q, McCartney JS, Parks J (2010b) Geotechnical issues in the design and construction of PMOBs. In: *UNSAT 2010*, Barcelona, pp. 1361-1367.

Candiani G, Silva ER, Moreira JML (2011) Methane balance in an experimental landfill cell. *Avances en Energías Renovables y Medio Ambiente*, 15:63-70 (in Portuguese).

Capanema MA, Ndanga E, Lakhout A, Cabral AR (2013) Methane oxidation efficiencies of a 6-year-old experimental landfill biocover. In: *Proceedings Sardinia, Fourteenth International Waste Management and Landfill Symposium*, S. Margherita di Pula, Cagliari, Italy. 2013.

Capanema MA, Cabana H, Cabral AR (2014) Reduction of odours in pilot-scale landfill biocovers. *Waste Management*, 34:770-779.

Chanton JP, Liptay K (2000) Seasonal variation in methane oxidation in a landfill cover soil as determined by an in situ stable isotope technique. *Global Biogeochemical Cycles*, 14:51-60.

Chanton J, Abichou T, Langford C, Spokas K, Hater G, Green R, Goldsmith D, Barlaz MA (2011) Observations on the methane oxidation capacity of landfill soils. *Waste Management*, 31:914-925.

Christophersen M, Kjeldsen P, Holst H, Chanton J (2001) Lateral gas transport in soil adjacent to an old landfill: factors governing emissions and methane oxidation. *Waste Management and Research*, 19:595-612.

Czepiel PM, Mosher B, Crill PM, Harriss RC (1996) Quantifying the effect of oxidation on landfill methane emissions. *Journal of Geophysical Research*, 101:16721-16729.

Delhomenie MC, Heitz M (2005) Biofiltration of air: A review. *Critical Reviews in Biotechnology*, 25:53-72.

Du R, Lu D, Wang G (2006) Diurnal, seasonal, and inter-annual variations of  $\text{N}_2\text{O}$  fluxes from native semi-arid grassland soils of inner Mongolia. *Soil Biology and Biochemistry*, 38:3474-3482.

Einola JKM, Sormunen KM, Rintala JA (2008) Methane oxidation in a boreal climate in an experimental landfill cover composed from mechanically-biologically treated waste. *Science of the Total Environment*, 407:67-83.

Fernandes JG (2009) Evaluation of biogas emissions in an experimental landfill. *Dissertação (Mestrado em Saneamento, Meio Ambiente e Recursos Hídricos) – Universidade Federal de Minas Gerais, Escola de Engenharia*, 101p. (in Portuguese).

Gallego E, Perales JF, Roca FJ, Guardino X (2014) Surface emission determination of volatile organic compounds (VOC) from a closed industrial waste landfill using a self-designed static flux chamber. *Science of the Total Environment*, 470-471:587-599.

Gebert J, Groenroeft A, Pfeiffer E-M (2011a) Relevance of soil physical properties for the microbial oxidation of methane in landfill covers. *Soil Biology and Biochemistry*, 43:1759-1767.

Gebert J, Groenroeft A, Miehl G (2003) Kinetics of microbial landfill methane oxidation in biofilters. *Waste Management*, 23:609-619.

Gebert J, Röwer IU, Scharff H, Roncato CDL, Cabral AR (2011b) Can soil gas profiles be used to assess microbial  $\text{CH}_4$  oxidation in landfill covers? *Waste Management*, 31:987-994.

Geck C, Scharff H, Pfeiffer E-M, Gebert J (2016) Validation of a simple model to predict the performance of methane oxidation systems, using field data from a large scale biocover test field. *Waste Management*, 56:280-289.

Humer M, Lechner P (2001) Microbial methane oxidation for the reduction of landfill gas emissions. *Journal of Solid Waste Technology and Management*, 27:146-151.

Huber-Humer M, Lechner P (2003) Effect of methane oxidation on the water balance of the landfill cover and the vegetation layer. In: *Proceedings Sardinia. Ninth International Waste Management and Landfill Symposium*, Cagliari, Italy, 2003.

Hudson N, Ayoko GA (2008) Odour sampling. Comparison of physical and aerodynamic characteristics of sampling devices: a review. *Bioresource Technology*, 99:3993-4007.

Jantalia CP, Santos HP, Urquiaga S, Boddey RM, Alves BJR (2008) Fluxes of nitrous oxide from soil under different crop rotations and tillage systems in the South of Brazil. *Nutrient Cycling in Agroecosystems*, 82:161-173.

Jugnia L-B, Cabral AR, Greer CW (2008) Biotic methane oxidation within an instrumented experimental landfill cover. *Ecological Engineering*, 33:102-109.

Kjeld A, Cabral AR, Gústafsson LE, Andradóttir HO, Bjarnadóttir HJ (2014) Microbial Methane Oxidation at the Fífholt landfill in Iceland. *Verktækni*, 30:31-36.



- Lakhouti A, Schirmer WN, Johnson TR, Cabana H, Cabral AR (2014) Evaluation of the efficiency of an experimental biocover to reduce BTEX emissions from landfill biogas. *Chemosphere*, 97:98-101.
- Lucernoni F, Rizzotto M, Tapparo F, Capelli L, Sironi S, Busini V (2016) Use of CFD for static sampling hood design: an example for methane flux assessment on landfill surfaces. *Chemosphere*, 163:259-269.
- Maciel FJ, Jucá JFT (2011) Evaluation of landfill gas production and emissions in a MSW large-scale experimental cell in Brazil. *Waste Management*, 31:966-977.
- Monteiro LS, Mota FSB, Silva WMB, Borges DA (2016) Emissions of gases through various types of materials used as cover layers of an experimental chamber filled with municipal solid wastes. *Journal of Environmental Science and Water Resources*, 05:24-31.
- Nagaraj TS, Lutenege AJ, Pandian NS, Manoj M (2006) Rapid estimation of compaction parameters for field control. *Geotechnical Testing Journal*, 29:497-506.
- Obersky L, Rafiee R, Cabral AR, Golding SD, Clarke WP (2018). Methodology to determine the extent of anaerobic digestion, composting and CH<sub>4</sub> oxidation in a landfill environment. *Waste Management*, 76:364-373.
- Perera MDN, Hettiaratchi JPA, Achari G (2002) A mathematical modelling approach to improve the point estimation of landfill gas surface emissions using the flux chamber technique. *Journal of Environmental Engineering and Science*, 1:451-463.
- Rachor IM, Gebert J, Gröngroft A, Pfeiffer E-M (2011) Assessment of the methane oxidation capacity of compacted soils intended for use as landfill cover materials. *Waste Management*, 31:833-842.
- Rachor IM, Gebert J, Gröngroft A, Pfeiffer E-M (2013) Variability of methane emissions from an old landfill over different time-scales, *European Journal of Soil Science*, 64:16-26.
- Rochette P, Eriksen-Hamel NS (2008) Chamber measurements of soil nitrous oxide flux: are absolute values reliable? *Soil Science Society of America Journal*, 72:331-342.
- Roncato CDL, Cabral AR (2012) Evaluation of methane oxidation efficiency of two biocovers: field and laboratory results. *Journal of Environmental Engineering*, 138:164-173.
- Rose JL, Mahler CF, Izzo RLS (2012) Comparison of the methane oxidation rate in four media. *Revista Brasileira de Ciência do Solo*, 36:803-812.
- Sadasivam BY, Reddy KR (2014) Landfill methane oxidation in soil and bio-based cover systems: a review. *Reviews in Environmental Science and Bio/Technology*, 13:79-107.
- Scheutz C, Kjeldsen P, Bogner JE, De Visscher A, Gebert J, Hilger HA, Huber-Humer M, Spokas K (2009) Microbial methane oxidation processes and technologies for mitigation of landfill gas emissions. *Waste Management and Research*, 27:409-455.
- Scheutz C, Pedicone A, Pedersen GB, Kjeldsen P (2011) Evaluation of respiration in compost landfill biocovers intended for methane oxidation. *Waste Management*, 31:895-902.
- Scheutz C, Pedersen RB, Petersen PH, Jørgensen JHB, Uendo IMB, Mønster JG, Samuelsson J, Kjeldsen P (2014) Mitigation of methane emission from an old unlined landfill in Klintholm, Denmark using a passive biocover system. *Waste Management*, 34:1179-1190.
- Stern JC, Chanton J, Abichou T, Powelson D, Yuan L, Escoriza S, Bogner J (2007) Use of a biologically active cover to reduce landfill methane emissions and enhance methane oxidation. *Waste Management*, 27:1248-1258.
- Trégourès A, Beneito A, Berne P, Gonze MA, Sabroux JC, Savanne D, Pokryszka Z, Tauziède C, Cellier P, Laville P, Milward R, Arnaud A, Levy F, Burkhalter R (1999) Comparison of seven methods for measuring methane flux at a municipal solid waste landfill site. *Waste Management and Research*, 17:453-458.
- UK Environment Agency (2010) Guidance on monitoring landfill gas surface emissions, LFTGN07 v2, Bristol, UK, 66p.